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BOSTON UNIVERSITY

GRADUATE SCHOOL

Dissertation

WAVE-LENGTHS OF CERTAIN LINES IN THE SECONDARY SPECTRUM OF HYDROGEN

by

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INTRODUCTION

The importance of the study of the secondary spectrum of Hydrogen, enhanced as it has been by the discoveries of Urey[1] and others,[2,3,4] has made it seem very desirable to secure values of the frequencies of the lines of the secondary or molecular spectrum with as great accuracy as possible. This desirability is occasioned in great part by the fact that the analysis of the molecular spectrum of Hydrogen shows the complete break-down of the Bohr theory which so excellently describes the atomic spectrum of this element. Furthermore, the great complexity of the spectrum of this most simple of molecules provides a most adequate means of checking any theory, for such theory must be competent to explain the spectra to the finest detail. Accordingly we herewith present a study of the molecular spectrum of Hydrogen, selecting a limited number of lines of a character suitable for best measurements, and between 4389 and 4512 A.U. Wave-lengths of these lines and the corresponding wave-numbers are presented in an endeavor to provide more accurate determinations for use in spectral analysis.

From the time of Hasselberg's work[5] and the first extensive photographic studies of Merton and Barratt[6] to the recent work of Finkelnburg[7], Bay and Steiner[8], and Gale, Monk and Lee[9], tables of Hydrogen wave-lengths in the many line spectrum have been published.[10] Discrepancies in these tables exist, and it is in the endeavor to provide a suitable set of Hydrogen standards, the use of which may permit a more exact band spectrum analysis and investigation of the isotopes of Hydrogen, that the following research is undertaken. A serious attempt was made to anticipate and provide for the elimination or reduction of the errors arising from mechanical and thermal sources, and it seems advisable to record rather fully the details of the

laboratory facilities, apparatus, and procedure.

DESCRIPTION OF THE LABORATORY.

The Rumford Committee Room at the Massachusetts Institute of Technology is located in the Eastman Spectroscopic Laboratory, a description of which appears elsewhere.[11] In particular, the room was designed to be as free from vibrational disturbances and thermal fluctuations as possible. The floor which supports the instrument piers is about twenty inches thick and floats free from rigid connection with the walls or adjacent floors. The room is approximately 40' x 8' x 8'. At the farther end, two massive concrete blocks, approximately 2'4" x 2'2" x 2'2", capped by a stone slab 4'9" x 2'4" x 2¼" support the grating, and at the other end of the room a single block of similar dimensions supports the combined slit and camera. A housing of wooden framework, covered with wall board, encloses the grating. The top of the housing is removable, and a means of access to the grating is also provided through the side. The camera box proper is approximately 2' square in cross-section, and extends between the grating housing and the slit pier. It is carried entirely on three wooden supports, no contact being made with the grating or slit pier. Access may be had to the interior of the box through the top, which is removable in sections. The grating housing, camera box, slit, and plate holder mechanism are made light tight by folds of black cloth. A smaller concrete pier supports the projection lens, and a heavy laboratory table near the entrance to the room supports the electrolysis apparatus, diffusion pump, and Hydrogen tube. Upon three I beams, supported free from contact with the isolated floor, a platform permits the experimenters to make necessary adjustments during exposures without danger of

vibrational disturbances. The room is divided into two sections by a light-tight partition erected on the platform between the small pier and the source table, with a heavily curtained doorway permitting passage between the sections. The smaller section, serving as a source room, is also provided with a stone sink, thus making the room available for the development of the photographic plates. The temperature of the grating room is thermostatically controlled. Two thermocouples are used, their reading being recorded on a Browne recording Potentiometer, one hung in the middle of the room and the other within the grating box. The circulation of air in the room was improved by the use of two fans, run at low speeds, heating elements being placed in front of each. One such element was operated by an automatic switch which also controlled the influx of cold air to the room. When the temperature fell the heating element would be cut in while the cold air supply would be cut off, and vice versa. In order to avoid overloading of the cooling capacity of the system a rheostat used to control the input to the transformer primary was placed entirely outside the spectroscopic laboratory. The transformer was located on the special platform in the source room. The temperature within the grating box was, during an exposure of three to four hours, kept within $\pm 0.05^{\circ}F$.

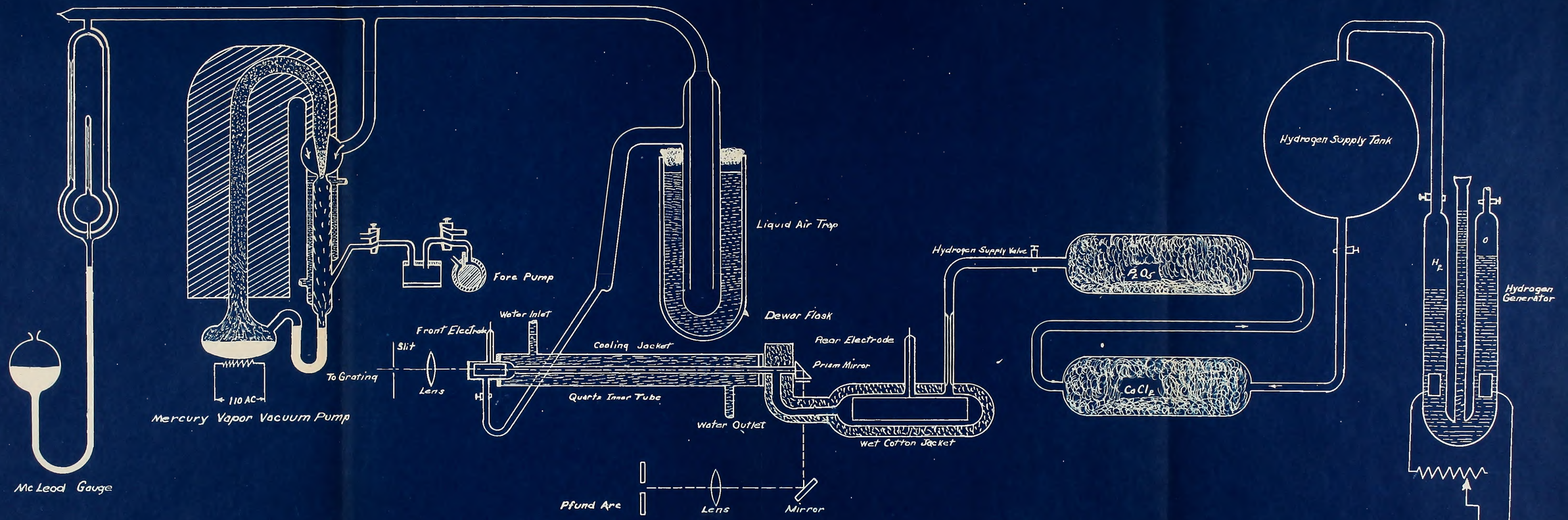
SOURCES

The Pfund[12] arc in air, as described by the International Astronomical Union[13], was operated at 220 volts and 4 amperes, a central zone less than 1.00 mm. in width of a 17 mm. arc being used. The upper pole, the cathode, was of Bessemer steel .25 in diameter. For the purpose of cooling and thus preventing the formation of a pendant bead of molten metal, a heavy brass

cylinder was fitted over the rod, which projected 2 or 3 mm. below the bottom of the cylinder. The anode consisted of a bead of iron oxide resting in a shallow cavity at the upper end of an iron rod $\frac{1}{2}$" in diameter. Babcock[14] mentions that the arc so constructed and used is very free from pole effect.[15]

The Hydrogen tube developed in this investigation embodies some of the features found in the discharge tubes presented by other investigators, in particular Finkelnburg[16], and Bay and Steiner[17]. The tube consists of a long quartz capillary 8mm. in diameter enlarged to 18 mm. at one end to contain one of the aluminum electrodes through which the discharge is viewed. A quartz window covers and seals this end of the tube. A side tube from this end leads via a ground seal to the evacuating system. The other end of the capillary is not expanded and is sealed off by a 90° prism. This arrangement makes it possible to line up the tube with the optical system with the minimum amount of effort[18] and provides opportunity of passing light from an exterior source, a secondary standard, say, down the tube so that both source and standard may be photographed simultaneously. A side tube leads from the capillary after a right angle bend, through a ground seal to a pyrex tube into which a cylindrical aluminum electrode is sealed. From this pyrex tube a small capillary, serving as a leak, leads to the hydrogen purifying tubes.

In cooling, the tube is provided with a pyrex water jacket extending from a point just behind the front electrode to the rear side tube of the capillary. In addition the rear electrode assembly is completely jacketed with absorbent cotton over which a steady flow of water is fed from holes in the side of a rubber connected with the water supply. This arrangement has made possible satisfactory cooling of the tube under all working conditions up to 175 m.a. at 30,000 volts.



ARRANGEMENT OF APPARATUS

PLATE 1.

It has been shown elsewhere,[19,20] that the presence of a metal in the Hydrogen discharge tube causes the relative enhancement of the molecular spectrum of Hydrogen, hence the interior of the tube was platinized by the use of Liquid Bright Platinum after the method of Kistiakowsky.[21] Essentially the process is to flow the liquid over the interior of the capillary and then to pass a current of air or oxygen through the tube while heating to redness in the flame of a blast lamp.

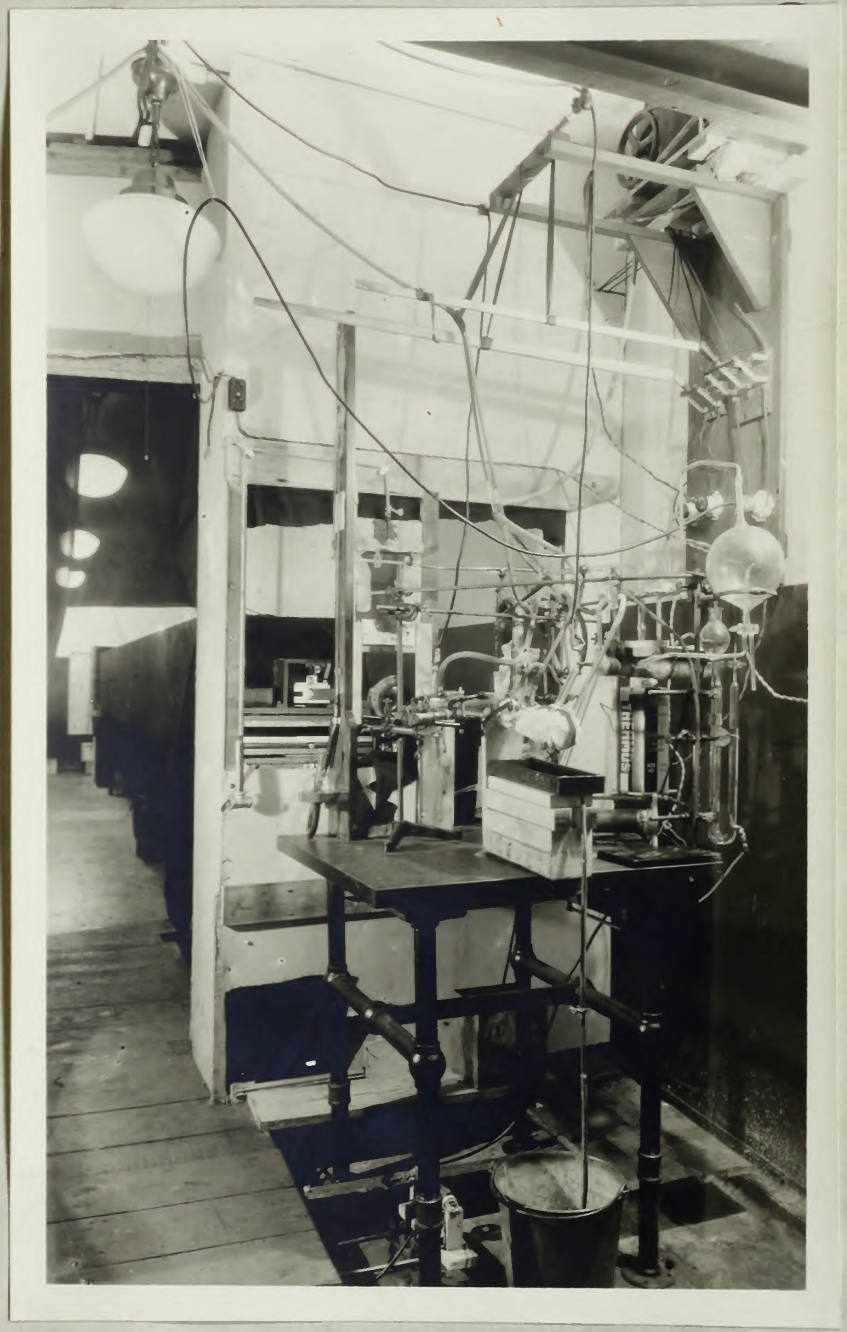
Some difficulty was experienced with the rear or external electrode which in the first construction was formed of thin sheet aluminum. After the tube had been in operation for a short time the leaks began to appear in the pyrex wall of the electrode chamber in the neighborhood of the edges of the cylinder. To avoid this apparent sandblast action it was found necessary to replace the thin metal electrode by a heavy closed cylinder whose edges were carefully smoothed off. No further trouble of the nature mentioned followed prolonged use of the tube after this change.[22]

OPTICAL SYSTEM.

The dispersive system consisted of a plane grating mounted as a Littrow. The arrangement of the various parts is indicated in Plate I. The grating, of Anderson construction, had a ruled surface 9.7 x 12.9 cm. and 12,542 lines to the inch. The dispersion in the second order was .88 angstroms per mm. Because of the inability of this grating to give a good line form it was found necessary after repeated tests to occult 4.5 cm. on one side of the ruling and 1.5 cm. on the other side. Under these conditions a good line form was obtained save in case of extremely heavy and therefore unmeasurable exposures.

The grating was mounted on a heavy steel member which rested by leveling screws on the alberlene slabs covering the grating piers. The grating was fixed in its holder by screws pressed against wax pads placed near the edges of the grating. Screws at the side of the grating holder permitted it to be rotated slightly in the plane of the ruled surface so that the lines of the grating might be made vertical; a screw at the rear allowed the grating to be given the necessary tilt. The holder was mounted on a vertical column arising from the steel base and could be rotated by a tangent screw thus enabling the desired region or order of the spectrum to be brought into position on the photographic plane.

The autocollimating lens used with this grating was a 30 ft. achromatic doublet with uncemented surfaces. It was mounted in front of the grating on a steel member attached by leveling screws which permitted the lens to be given the proper orientation. Ordinarily the screws would be used to place the axis of the lens so that it would intersect at its mid-point a line drawn from the center of the slit to the center of the photographic plate or plate-holder. Under these conditions however there appear, as has been pointed out by Buisson and Fabry[23], three images of the illuminated slit formed by reflection at the curved surfaces of the lenses. The images thus formed produce a great amount of background illumination at the plate-holder and to avoid this disturbance the remedies suggested by the authors mentioned above were undertaken, as follows: Two of the images are virtual. One of these is eliminated by tilting the lens slightly till the image just vanishes as viewed from the top and the bottom of the plate-holder. The other image is occulted by covering the lens with a narrow strip of black paper. Finally, the third image, which is real, is covered at its focus by a small pellet of wax at-



tached to a thread stretched across the interior of the camera box. When these operations was carried out the spectra appeared against a black background.

Pinned at a convenient position on the slit and plate-holder pier, a heavy steel plate supports between ways parallel to the axis of the instrument a massive rectangular steel box open at the front and rear. The plate-holder which is mounted within this box is capable of a horizontal rotation about a vertical axis at approximately the center of the plate. The proper tilt having been obtained by such rotation, the plate-holder may be clamped in position by a locking nut on the vertical screw about which it turns.

In use the instrument accomodates a plate 24" x 2" which rests against flexible brass strips which may be bent and held to any desired curve by means of adjusting screws.

The slit mechanism is mounted in a housing bolted to the top of the plate-holder box. The slit carriage is capable of a longitudinal motion parallel to the axis of the instrument so that the slit may be placed in the plane of the center of the plate. The slit may be rotated in a plane normal to the axis of the instrument so that its jaws may be made vertical; or it may be rotated about a vertical axis so that the slit jaws may be set normal to the optical axis.

The arrangement of the complete apparatus is evident from Plates I and II. It will be noted that due to lack of space the Pfund arc has been placed at one side, and light from the arc which is focussed by means of a lens on the front end of the tube is brought to this position by means of reflection from an aluminum-surfaced mirror[24] and the right-angled prism with which the tube is sealed.

METHODS OR PROCEDURE.

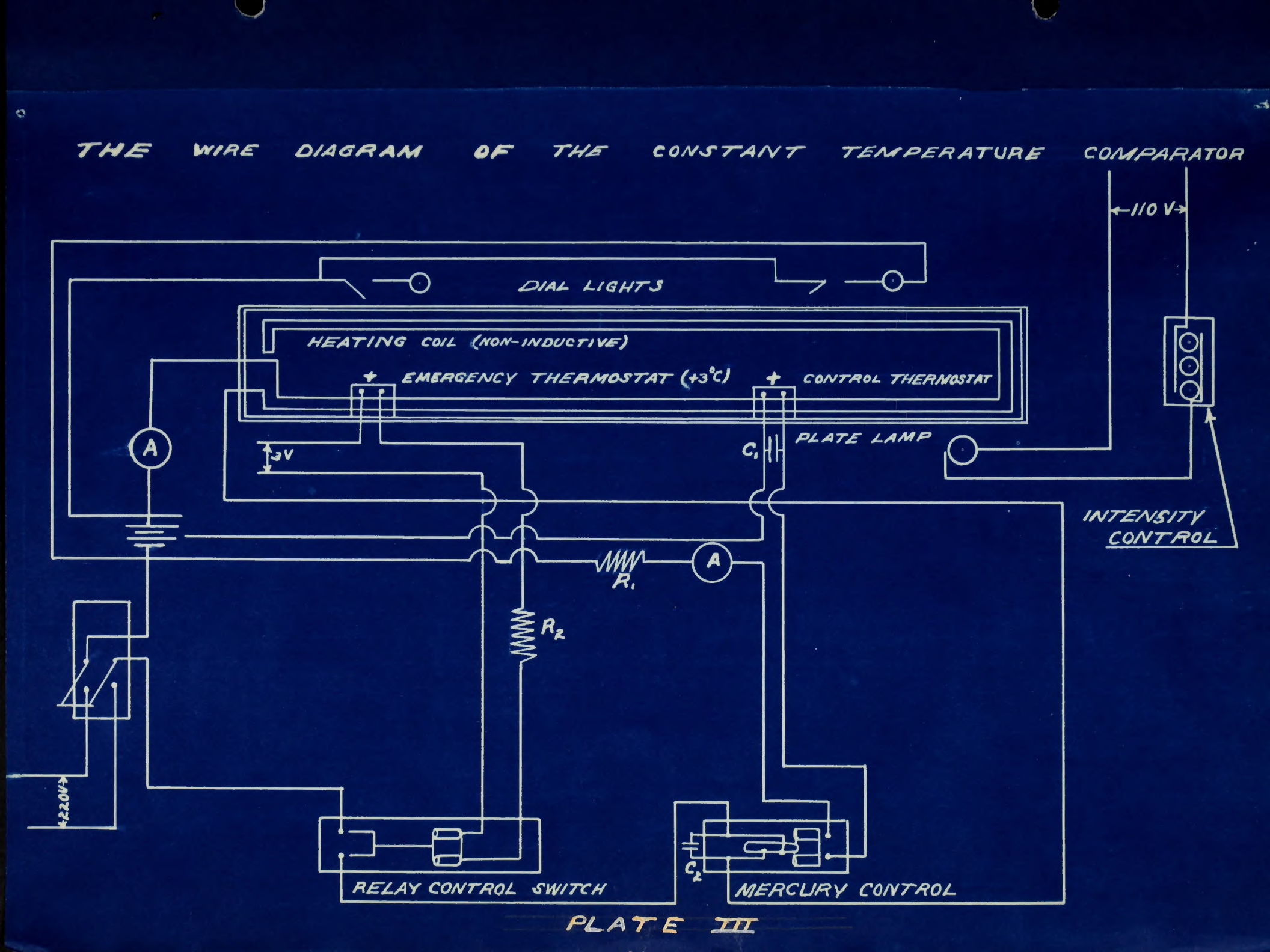
The various parts of the apparatus described were arranged as indicated in the diagram of Plate I. Their alignment presented no great difficulty owing to the fact that the construction of the discharge tube permitted the experimenter sight along the entire optical axis from the source to the grating. After rough leveling and lining up of the various parts a small light source (flash lamp bulb) was placed as nearly as possible on the axis of the system and directly in front of the autocollimating lens. With the slit wide open and a small stop over its mid-portion the table bearing the tube was oriented till the source was viewed centrally throughout the length of the 8 mm. capillary. The light on passing through the tube was reflected by the right angle prism to the mirror, through the lens, and thence to the position of the arc. This beam of light afforded a means of putting the mirror, lens, and the source in approximate position. The exact position of mirror and lens was determined by viewing their images as formed by the projection lens with a small magnifier and arranging their center on the axis of the tube. Cross hair on the lens and the mirror enabled this to be done with great ease. The adjustment was verified by lighting the tube and noting that with the projection lens removed the shadow of the slit fell central on the autocollimating lens. The Pfund arc was focussed by a small achromat after two reflections on the end of the capillary next the prism. The length of the arc was 17 mm. and the magnification approximately 10 x 1. Under these conditions a horizontal central section of the arc less than 1 mm. in height was used.

The Hydrogen discharge tube was operated at 30,000 volts and 175 m.a.

from the secondary of a 5 kw. Acme transformer. A large rheostat limited the power input to the primary. Before every exposure of a plate the tube was operated until with a continuous flow of Hydrogen the secondary spectrum appeared strong and the Balmer series weak. By adjustment of the rate of production of Hydrogen in the electrolytic generator and the manipulation of stopcocks it was found possible to maintain this optimum condition throughout the period of exposure. Frequent examinations of the discharge were made with a direct vision spectroscope while the spectrum was being photographed.

In order to increase the intensity of the tube on the slit a small mirror was lightly pressed by a spring against the surface of the mirror so that the tube light emitted to the rear would be reflected down the tube. This mirror was removed when the arc exposure was being made or when the direct vision spectroscope was being used. To reduce errors to a minimum the arc exposure, which due to greater intensity was considerably less than that of the tube, was divided into at least five parts spaced at equal intervals throughout the hydrogen exposure. The grating was turned up to the second order on the right, in which order the maximum brightness was obtained and set for the region between 4000 A and 4500 A. The spectroscope was focussed visually and then photographically for the center of the plate, and then by photographic methods the other parts of the plate were brought in to the focal plane of the instrument.

The plates used in this research were supplied by the Kodak Research Laboratories and were their Type 1-0.[25] These plates were of high speed but only of medium contrast. The emulsion was coated on thin glass and all plates were backed. The plate size was 2" x 22". The exposures for the Pfund arc were from 15 to 60 minutes and for the Hydrogen discharge 3 hours.


THE WIRE DIAGRAM OF THE CONSTANT TEMPERATURE COMPARATOR
110 V
DIAL LIGHTS
HEATING COIL (NON-INDUCTIVE)
+ EMERGENCY THERMOSTAT (+3°C)
+ CONTROL THERMOSTAT
A
3V
C1
PLATE LAMP
INTENSITY CONTROL
A
R1
R2
220V
RELAY CONTROL SWITCH
C2
MERCURY CONTROL
PLATE III

Development of the plates took place immediately after exposure in a contrast developer D-11.[26] Plates were developed for 6 minutes at a temperature of 65°F. This time yielded the maximum contrast possible without the appearance of fog. The plates were fixed in an acid-hardener fixing solution, washed and carefully squeegeed and dried.

The plates were measured on the Société Génévoise measuring machine of the Physics Department of Boston University. This instrument is elaborately housed in a constant temperature cabinet, and provided with automatic temperature control. A sketch of the control system appears in the diagram of Plate III. Three plates were measured, two runs being made on each. The iron lines selected as standards were measured five times on each run, whereas the hydrogen lines were measured twice. For the second of the two runs the plate was reversed in its position on the comparator so that the lines would be approached from a different direction in making a setting and so that a different position of the comparator screw would be used.

In reducing the measurements the wave lengths of six standard iron lines between the standards of standards were first calculated and the difference between the wave-lengths thus obtained and the accepted values were plotted on a large scale against wave-length. A smooth curve was fitted to the points so plotted. Two such curves were drawn for each plate.

The hydrogen wave lengths were calculated by interpolation or extrapolation from the standards of standards and then corrections taken from the appropriate curve as described above were applied. The wave lengths so computed appear in the table following.

WAVE LENGTHS IN THE SECONDARY SPECTRUM OF HYDROGEN

Wave Lengths in A.U. (Roots)							Wave number (in vacuo)	Mean, [7] Finkelnburg and Gale, Monk,[9] and Lee	Mean, Roots - Mean, Finkelnburg and Gale, Monk, and Lee
	Int.	Plate No. 36	37	38	Mean	Ave. Dev.			
4389	0^2	080	083	084	082	002	22777.43	090	-8
4390	0	899	901	907	902	3	22767.99	899	3
4391	0^4	732	736	737	735	2	22763.67	732	3
4392	0^4	088	090	100	093	5	22761.81	097	-4
4410	1	486	481	489	485	3	22666.95	483	2
4410	1	613	617	615	615	1	22666.24		
4412	8_3	248	255	254	252	3	22657.82	253	-1
4412	0^3	476	480	485	480	3	22656.65	478	2
4413	0^3	494	498	498	497	2	22651.43	502	-5
4414	2	216	220	220	219	2	22647.73	213	6
4415	0	994	012	007	004	7	22643.70		
4416	0^4	219	225	228	224	3	22637.44	219	5
4417	4	335	338	341	338	2	22631.74	340	-2
4419	0^2	485	497	492	491	4	22620.71	492	-1
4420	0^3	300	310	302	304	4	22616.55	305	-1
4422	0^4_3	986	993	988	986	2	22602.84	989	-3
4423	0^3	246	249	252	249	2	22601.49	251	-2
4425	0^3	150	157	153	153	2	22591.77	150	3
4425	0	896	905	905	902	2	22587.94	898	4
4434	0^4	989	989	991	990	1	22541.66	986	4
4438	0^4	633	630	644	636	5	22523.14	633	3
4441	0^4	413	407	420	413	4	22509.06	412	1
4442	0^4	860	859	865	861	4	22501.72		
4444	0	219	224	217	220	3	22494.84	216	4
4445	5	249	253	249	250	2	22489.63	245	5
4447	0	930	930	932	931	1	22476.07	930	1
4449	1_4	915	923	921	920	3	22466.03	916	4
4450	0^4	835	831	841	836	4	22461.41	816	20
4452	0^4	778	776	776	777	1	22451.62	771	6
4453	0^4_3	161	160	159	160	1	22449.68	151	9
4456	0^3	669	675	679	674	4	22431.99	657	17
4456	0_3	862	864	865	864	1	22431.03		
4457	0^3	037	039	038	038	1	22430.15	037	1
4458	0^3	749	749	758	753	4	22421.53		
4460	10	969	971	968	969	1	22410.39	968	1
4464	0^4	228	231	231	230	1	22394.02	228	2
4467	6_4	146	148	142	145	2	22379.40	143	2
4471	0^4	961	965	959	962	2	22355.30	957	5
4474	3	258	259	259	259	1	22343.82	265	-6
4477	2_4	073	071	073	072	1	22329.78	072	0
4478	0^4	985	993	981	986	4	22320.24	984	2
4481	0^4	257	255	262	258	3	22308.92	268	-10
4481	0^4	907	915	912	911	3	22305.67	919	-8

WAVE LENGTHS IN THE SECONDARY SPECTRUM OF HYDROGEN

Wave Lengths in A.U. (Roots)							Wave number (in vacuo)	Mean, Finkelnburg and Gale, Monk, and Lee	Mean, Roots - Mean, Finkelnburg and Gale, Monk, and Lee
	Int.	Plate No. 36	37	38	Mean	Ave. Dev.			
4485	0^3	837	842	839	839	002	22286.14		
4486	5	082	078	083	081	2	22284.94	089	-8
4487	6	813	816	817	815	2	22276.33	819	-4
4490	5	451	457	452	453	2	22263.26		
4493	3	690	695	689	691	2	22247.35	691	0
4497	0^4	102	104	105	104	1	22230.32	098	6
4497	0^4	577	578	585	580	3	22227.96	573	7
4498	7	110	109	105	108	2	22225.36	108	0
4498	4	525	532	533	530	3	22223.27	523	7
4501	3	966	966	965	966	1	22206.31	960	6
4505	4	639	642	640	640	1	22188.20	633	7
4510	1	917	922	913	917	3	22162.25	901	16
4511	1	710	716	709	712	3	22157.33	698	14
4512	0^2	882	---	874	878	3	22152.61	874	4

Note: Intensities are given in the second column and are eye estimates. The symbol "0^4" signifies the weakest line measured and "10" the most intense. Not all lines on the plate were good enough for satisfactory settings and such lines were not measured. The wave numbers in the eighth column were obtained by the use of Kayser's Tabelle der Schwingungszahlen.

DISCUSSION

Examination of the results as given in the table reveals that the mean average deviation in our measurements is .003 A.U. The most frequent deviation is .002 A.U., and average deviation equal to or greater than .005 A.U. occur in only three instances.

In the last column of the table we have compared the mean of the results of Finkelnburg and Gale, Monk and Lee with our data. The differences for 50 lines show that our results are systematically higher than the mean of these workers by .002 A.U. A comparison with the separate determinations of Finkelnburg and Gale, Monk and Lee show that on 55 lines the measurements of the former are systematically greater than ours by .001, whereas on 56 lines those of Gale, Monk and Lee are less by .004.

The region we have investigated contains four lines measured by Gale, Monk and Lee by interferometer methods. Comparison of those measurements with ours is given in the following table.

Wave Lengths in A.U.

Roots (grating)		Gale, Monk and Lee (interferometer)	R - G, M & L
4412	252	253	-001
4460	969	965	004
4498	108	108	000
4490	453	451	002

The average R - G.M.L. difference is here $<.002$ A.U.

From the limited number of lines available it may be seen that our grating measurements are in somewhat better agreement with the more precise interferometer determination of Gale, Monk and Lee than with their grating determinations. This may be due in part to the fact that their grating photographs were made with a dispersion of 2.63 A.U./mm. as against .88 A.U./mm. used in this research. It may be remarked in this connection that

Finkelnburg's grating measurements, with which we are in better agreement, were made with a dispersion of 1.98 A.U./mm.

CONCLUSION

A determination of the wave-length of fifty-seven lines in the spectrum of molecular hydrogen lying between 4389 and 5412 A.U. was made. A tube of new design was used as the source, and the secondary spectrum was photographed in the second order of a 30 foot Littrow mount with an average dispersion of .88 A.U. per mm. Great care was taken to avoid thermal and mechanical disturbances during the exposures. Three plates were exposed and selected lines were measured. The results as compared with those of certain other recent investigators show small systematic deviations.

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VITA

The author, Yale Kirby Roots, was born in Milford Center, Ohio, on February 3, 1894, the son of Albert Bartlett and Dorcas B. Erwin Roots. He graduated from the Irwin, Ohio, High School in 1911, and received the degree of Bachelor of Science in 1922 from Wooster College, and of Master of Science in 1926 from New York University. From 1923 until June 1930, he was an instructor in the Physics Department of New York University. Since that time he has been head of the Department of Physics and Mathematics at Findlay College. He holds membership in the following scientific societies: American Physical Society, American Association for the Advancement of Science, Mathematical Association of America, Ohio Academy of Science, Optical Society of America, Royal Photographic Society, and Sigma Pi Sigma. He is joint author, with F. A. Jenkins and Robert S. Mulliken, of "The Red CN Band System", published in the Physical Review, vol. 39, pp.16-41, January, 1932.

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